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# Air Pollution: Ground-Based Sensing of Source Emissions

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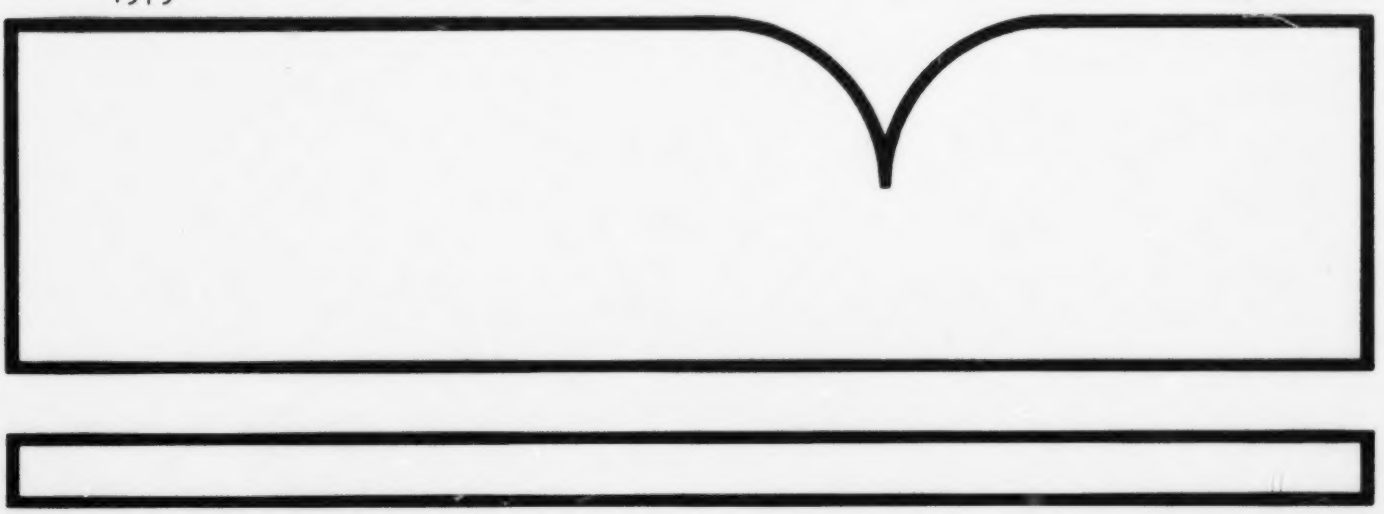

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16. ABSTRACT

Some types of gaseous pollution sources, particulary extended area industrial complexes and those producing hot combustion products, cannot be monitored adequa tely with conventional point sampling methods. To aid in characterizing emissions from and in developing remote sensing methods for application to such sources a high-resolution (0.06 $cm^{-1}$) infrared spectrometer system has been installed in a van. This system can be used with a remote light source to make longpath absorption measurements in the atmosphere or can be used in a single-ended mode to obtain emission spectra of hot gases. Initial measurements were made across a fertilizer plant gypsum pond, hydrogen fluoride was measured in the 10-50 ppb range. Measurements at an oil refinery showed propylene, ethylene, methanol, other hydrocarbons, sulfur dioxide, and carbon monoxide in ppb to ppm levels. Jet engine emissions were also evaluated. The measurement system is described and data from the above three measurement programs are presented.

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# 3 AIR POLLUTION: GROUND-BASED SENSING OF SOURCE EMISSIONS

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## I. REMOTE MEASUREMENT OF SOURCE EMISSIONS

In early 1968, the National Air Pollution Control Administration (later to become a part of the EPA) recognized the need for advanced instrumentation that could identify and measure the concentration of gaseous pollutant emitted by various types of sources. It was determined that an infrared spectrometer system capable of making (1) long-path (about a kilometer) absorption measurements in the atmosphere and (2) single-ended remote emission measurements of stack effluents would be a particularly suitable instrument for characterizing the emissions from various sources. Specifically, such measurements would require no prior knowledge of anticipated species, no sample handling, and the spectral signatures of interfering species as well as particular pollutants could be determined simultaneously. It was anticipated that these later measurements would provide background data for the development of simple pollutant-specific infrared measurement systems.

Such an instrument, called the ROSE (Remote Optical Sensing of Emissions) system, was developed for EPA by General Dynamics/Convair under the direction of Dr. Claus Ludwig (Streiff and Claysmith, 1972; Streiff and Ludwig, 1973). The original system utilized a blackbody radiation source, Dall–Kirkham telescopic optics, a quarter-meter linear-



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wavenumber-drive monochromator, and a cryogenically cooled Hg:Ge detector to cover the 800–5000 $cm^{-1}$ spectral region. For long-path measurements, this system provided a spectral resolution of about 1 $cm^{-1}$. For single-ended emission measurements of industrial stack effluents (temperature on the order of 400°K) the highest achievable resolution compatible with a reasonable signal-to-noise ratio was about 10 $cm^{-1}$. Despite these resolution limitations, a number of successful measurements were carried out on the CO fundamental band at 2145 $cm^{-1}$ and on the $SO_2$ combination band at 2500 $cm^{-1}$. However, it was realized that a significant improvement in spectral resolution was needed to allow successful implementation of the long-path absorption/emission measurement concept. Accordingly, in late 1975 it was decided to replace the grating monochromator and its associated detector and electronics with a Fourier transform interferometer system. For a complete discussion of various remote sensing methods and applications, the reader is referred to Herget and Connor (1977) and Ludwig and Griggs (1976).

## II. THE ROSE SYSTEM

A schematic of the ROSE optical system is shown in Fig. 1. The light source–telescope system used for the long-path absorption measurements is shown in Fig. 2. The $f/5$ scope is of Dall–Kirkham configuration

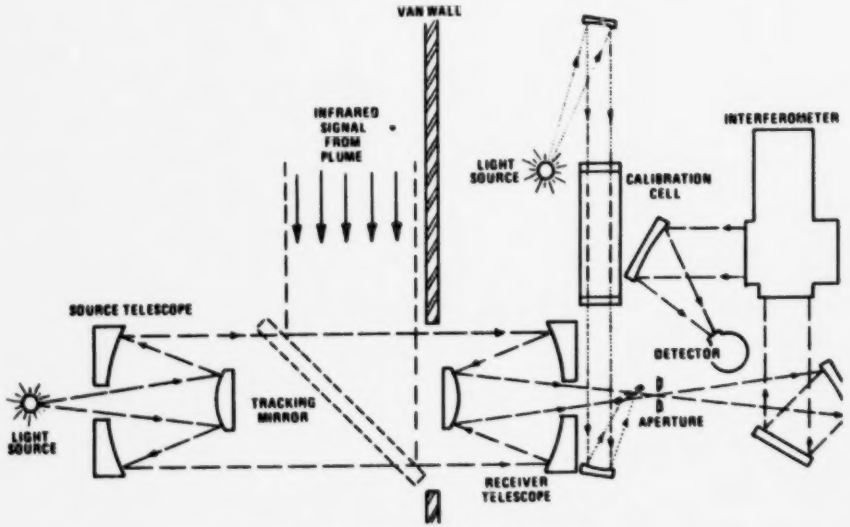


**Fig. 1.** The ROSE optical system.



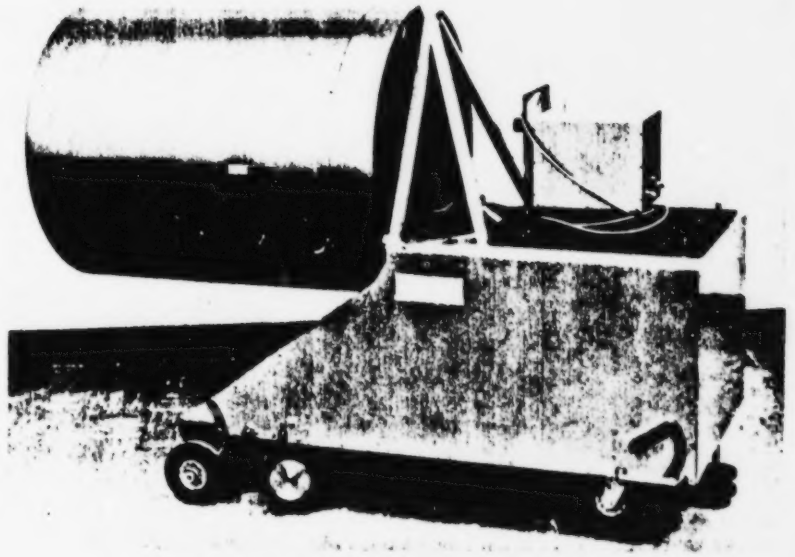

**Fig. 2.** The ROSE system light source telescope.

with a 30-cm-diam. primary mirror. Presently the light source is a 1000 W quartz–iodine lamp. This lamp provides considerably more near-infrared energy that the 1500°K blackbody previously used (shown in Fig. 2) and in wavelength regions where the quartz envelope is opaque, its thermal emission provides sufficient energy. In addition, the intense visible energy from this source aids system alignment. Generally, the light source and telescope system is installed in the back of a pickup truck and driven to a desired location; a small generator powers the light source if local power is not available.

The remainder of the ROSE system is installed in a 28-ft van. A telescope identical to that described above collects energy from the remote light source directly through a port in the side of the van, as indicated in Fig. 1. To measure the radiation signal from stack effluents, an elliptically shaped flat mirror (called the tracking mirror in Fig. 1) reflects energy through the port into the telescope. The tracking mirror is mounted on a platform attached to the side of the van (Fig. 3). The receiver telescope focuses energy (in either mode of operation) at the interferometer aperture, which is adjustable for compatibility with desired spectral resolution. The

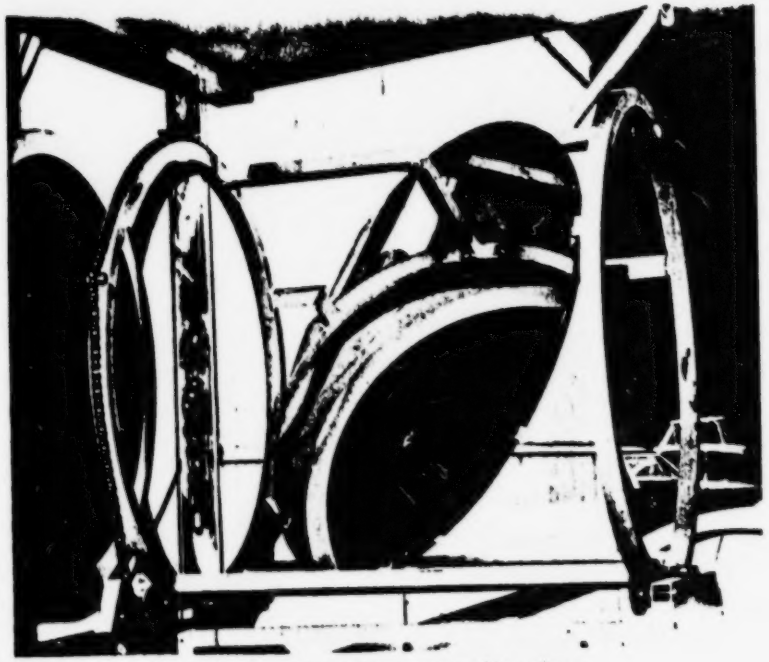

Fig. 3. The ROSE system tracking mirror.

interferometer and peripheral equipment comprise a standard Nicolet Instrument Corporation Model 7199 FT-IR system configured to fit into the van. Major components of this system consist of a computer with 40K memory, dual-density disk with 4.8 million, 20-bit word capacity, teletype, paper tape reader, oscilloscope interactive display unit, and a high-speed digital plotter. Maximum spectral resolution achievable with this system is 0.06 $cm^{-1}$.

The interferometer itself is mounted on the telescope support structure. All other systems (except the plotter) are arranged in two 19-in. relay racks. The general layout of the van is shown in Fig. 4, and a major portion of the interferometer system is shown in Fig. 5. Two beam splitters, KBr and $BaF_2$, are available for use in the interferometer. A dual-element, sandwich-type detector is mounted in a liquid nitrogen Dewar. For the 1800–6000 $cm^{-1}$ region InSb is used, and HgCdTe is used from 600 to 1800 $cm^{-1}$; the two regions are scanned separately.

For the long-path absorption measurements, initial alignment of the telescopes is achieved by visually boresighting each at the other. Final



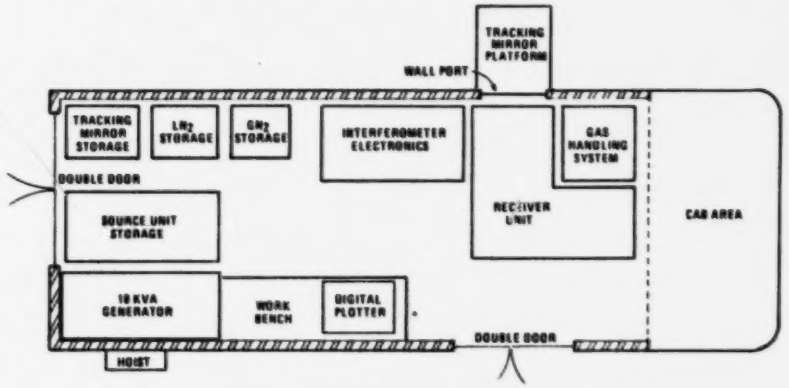


Fig. 4. Layout of ROSE system in van.

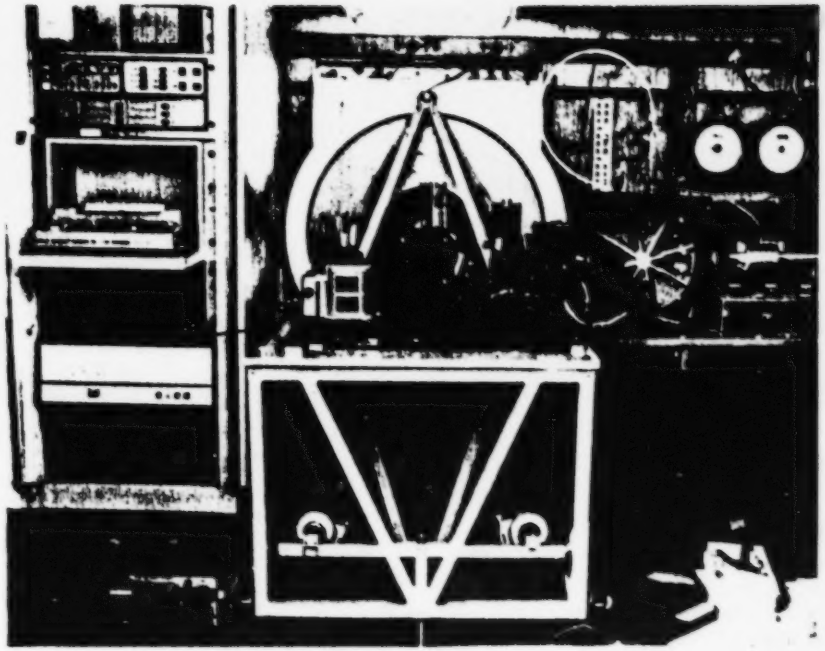

Fig. 5. The ROSE system.

alignment is made by maximizing source signal level at the interferometer aperture with the assistance of two-way radios. For stack emission measurements, alignment using the tracking mirror is obtained by boresighting and by observing the image projected by the telescope. The tracking mirror can be adjusted manually from inside the van about the telescope axis and a vertical axis. This alignment system is sufficiently precise to allow manual tracking of the sun for solar absorption measurements.

Auxillary equipment carried in the van includes a weather station for recording wind velocity and temperature and a laser range finder for measuring pathlengths. For stability in the long-path configuration four jacks are lowered from the van chassis to the ground. The interferometer itself is leveled within the van. Power for the ROSE system, including that needed for heating or air conditioning, is supplied by a 10 kW generator. For data collection, the generator is lowered from the van to the ground using an electrically operated winch. This procedure is necessary to avoid electrical and mechanical interference with the operation of the interferometer. During transportation of the van to a field site, two precautions are observed: the beam splitter is removed from the interferometer and the disk drive head is clamped back away from the disk. After reinsertion of the beam splitter, the interferometer can be realigned in a matter of minutes. An initial setup of the entire system requires two to three hours. Subsequent setups in the same vicinity require about one hour under normal conditions.

## III. SYSTEM CALIBRATION

The calibration of the ROSE system for long-path measurements follows the conventional procedure of obtaining the spectra of known concentrations of desired species (broadened to atmospheric pressure) contained in a sample cell (shown in Fig. 1). Usually some preliminary spectra are obtained prior to a field trip. The final calibrations are made after the field trip in order to match instrument resolution and sample absorption with field measurement conditions. A vacuum transfer system with appropriate gauges is used in filling the cell to desired pressures. Cells ranging in length from 1 to 200 cm are available.

In order to obtain an intensity calibration of the emission spectra due to hot gases, a blackbody source operated at known temperature is caused to fill the interferometer field-of-view and its spectrum recorded. System software has the capability of generating the Planck blackbody radiation function for any desired blackbody temperature. By dividing the blackbody spectrum by the appropriate Planck function, an instrument



response function is obtained. An emission spectrum obtained from a plume or flame (that also fills the interferometer field of view) can be divided by the instrument response function. This result can then be plotted in terms of spectral radiance (watts centimeter$^{-2}$ steradian$^{-1}$ wave number$^{-1}$). If the temperature of the hot gas plume can be determined, then a Planck blackbody function can be generated at the temperature of the plume, divided into the spectral radiance curve, and this result plotted to yield spectral emissivity as a function of wave number. (All of these operations are carried out using standard system software.) Then, if the spectral absorption coefficient of the species in question is known as a function of temperature, the gas concentration can be calculated. The basic equations relating these quantities are:

$$\tau(\nu) = e^{-K(\nu,T)CL}$$

$$\epsilon(\nu) = N_P(\nu)/N_{BB}(T_P,\nu)$$

$$\tau(\nu) = 1 - \epsilon(\nu)$$

where $\nu$ is the wave number (cm$^{-1}$), $\tau(\nu)$ the transmittance of source energy, $K(\nu,T)$ the spectral absorption coefficient [(atm cm)$^{-1}$ (ppm meters)$^{-1}$ and (molecules cm$^{-2}$)$^{-1}$ are typical units], $C$ the concentration, $L$ the pathlength (source to receiver distance or plume thickness), $\epsilon(\nu)$ the plume emissivity, $N_P(\nu)$ the spectral radiance of the plume, and $N_{BB}(T_P, \nu)$ the spectral radiance of a blackbody at the plume temperature. The determination of the plume temperature can be a complex problem in itself and will not be discussed here.

## IV. MEASUREMENT PROGRAMS

One of the principal aims in the use of the ROSE system is to characterize (determine species identity and concentration) the gaseous pollutants emitted from various types of sources, and in particular, sources that are difficult to characterize by conventional point sampling methods. These types of sources fall into two general categories: (1) extended area sources where the pollutants may be emitted from many locations (leaks, stacks, ponds, etc.) within a particular area; and (2) point sources of warm ($T > 400$°K) combustion products such as gas turbines and flares. Three sources typical of the above two categories were selected for the first field measurement programs involving the ROSE system. Fortunately, it was possible to select sources which both were of particular importance to Agency needs and also would allow test of the ROSE system under a variety of field conditions. These three programs are described below.

### A. Gypsum Ponds

The first field use of the ROSE interferometer system was at a phosphate fertilizer plant gypsum pond. A series of these ponds are used at fertilizer plants for waste water treatment. The ponds, which are generally rectangular in shape with boundary dimensions as long as a kilometer, are particularly suitable for long-path measurements. The particular environmental problem presented by these ponds is that they give off gaseous fluorides. In past studies using wet chemistry sampling methods, it had been possible to measure only total fluorides. Analysis of the pond chemistry indicates that expected gaseous fluorides would be $SiF_4$ and/or HF. Thus a study was undertaken at the C.F. Industries fertilizer plant near Bartow, Florida, to determine specifically which gaseous fluorides are emitted from the ponds.

A series of measurements were made at various locations around several ponds with pathlengths ranging from 500 to 1000 m. Typical spectra obtained are shown in Fig. 6. The upper spectrum was taken over a 900 m path at a location known to be free of HF. The middle spectrum was taken over an 860 m path across a gypsum pond. Both spectra were taken with a resolution of 0.125 $cm^{-1}$. Using system software, these spectra were expressed in absorbance, subtracted, and the result converted back to transmittance. This result is shown in the lower spectrum. The subtraction eliminates the effect of the weak atmospheric absorption seen in the upper spectrum at 4173.9 $cm^{-1}$. Using an HF line strength of $3.48 \times 10^{-19}$ $cm^{-1}$ (molecules $cm^{-2})^{-1}$ (Randall, 1975) and a half-width of 0.04 $cm^{-1}$ (Meredith and Smith, 1974), the HF concentration was determined using the equivalent-width method. The calculations were carried out with an existing computer program (Burch and Gryvnak, n.d.). For the HF line shown, the path-averaged concentration was determined to be 45 ppb. (It was not possible to calibrate the HF spectrum with the sample cell method since our gas handling system is not resistant to HF.) Absorption due to the $SiF_4$ fundamental band centered at 1031.5 $cm^{-1}$ could not be detected. Calibration spectra indicated that 2.0 ppb of $SiF_4$ would have produced about 2% absorption over an 860 m path, and this value is taken as a reasonable lower sensitivity limit.

### B. Jet Engines

In order to obtain some basic data on jet engine emissions as well as to learn how the interferometer would perform in a high-noise-level environment, a series of measurements were made at Tyndall Air Force Base, Florida, in cooperation with personnel of the Air Force Civil Engineering Center. Three measurement configurations were selected: (1) to measure



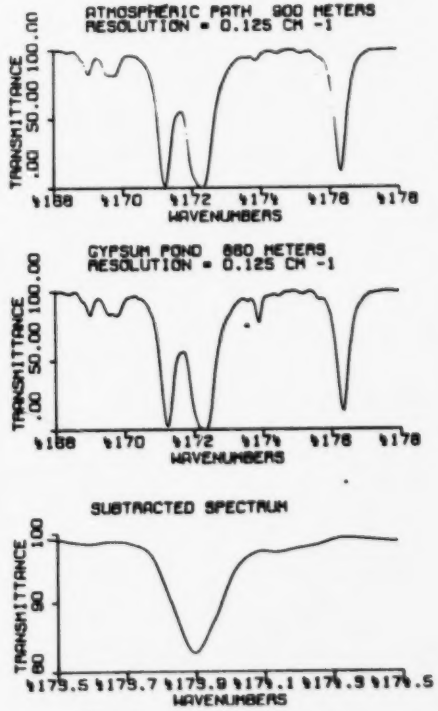


**Fig. 6.** Gypsum pond spectra.

the absorption of the exhaust of a single jet engine, the light source and ROSE van were located so as to provide an optical path perpendicular to the engine axis and 10 m down stream of the nozzle exit (this particular engine was on a test stand), with the van being about 80 m from the engine; (2) to measure the emission signal from a single engine, the receiver telescope was aimed at the nozzle exit plane of an engine being operated in a tethered aircraft, with the van being about 60 m from the aircraft; (3) for a long-path absorption measurement the ROSE van and light source were separated by 900 m and the optical path was parallel to a taxiway.

Measurements were made in (1) and (2) with the engines operated at idle, military, and afterburner power. During the measurements on the exposed engine, (1), it was found that for military and afterburner power levels the interferometer operation was severely affected by acoustical noise and the data were useless. For the engine in the aircraft, (2), only

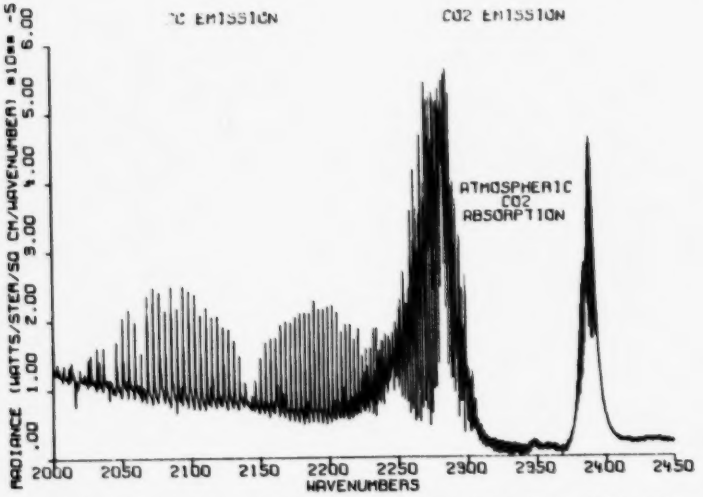


**Fig. 7.** Jet engine emission spectrum (idle power); resolution, 0.25 $cm^{-1}$.

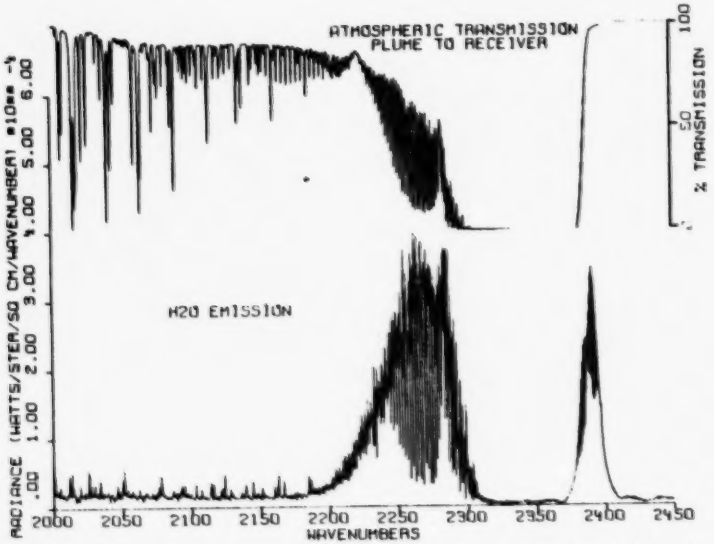


**Fig. 8.** Jet engine emission spectrum (military power); resolution, 0.25 $cm^{-1}$.



afterburner operation affected instrument performance. Although time did not permit additional measurements, it was felt that if the ROSE van were located possibly 200 m from the engines, then good spectra could be obtained under any operating conditions.

Samples of emission spectra obtained in the 2140–2410 $cm^{-1}$ spectral region at idle and military power are shown in Figs. 7 and 8. Noteworthy spectral characteristics are CO emission at idle and lack of CO emission at military power. Absorption spectra obtained at idle power are shown in Fig. 9. The calculated concentrations, based on a plume diameter of 10 m, are given in Table 1. During the time slot allotted to the long-path measurements, there was not much aircraft activity due to a number of rain squalls. The spectra obtained did show CO at about 0.6 ppm and also the hydrocarbon continuum with an equivalent hexane concentration of about 0.09 ppm.

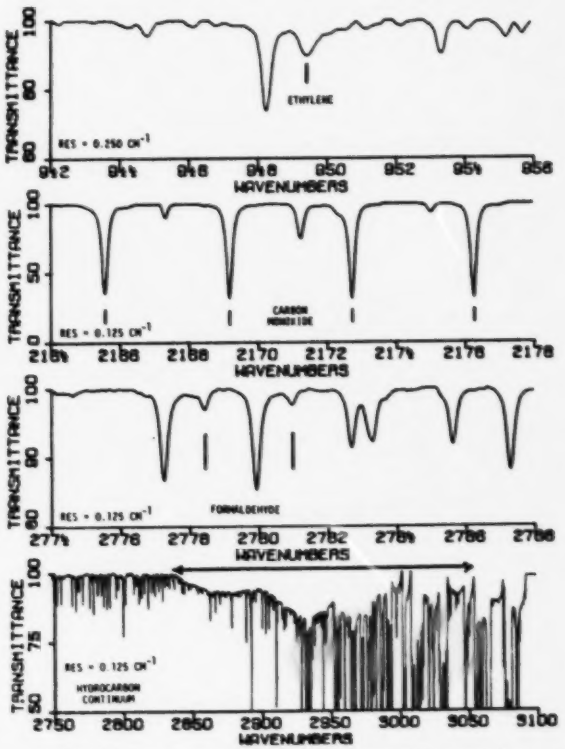


**Fig. 9.** Jet engine absorption spectra (idle power).

**TABLE 1**

Species Concentrations in Jet Engine Exhaust (10 m path)

| Species | $\nu$ ($cm^{-1}$) | $K(\nu)$ (ppm meter)$^{-1}$ | Reference for $K(\nu)$ | $C$ (ppm) |
|---|---|---|---|---|
| Ethylene | 949.4 | $3.3 \times 10^{-3}$ | This work | 3.2 |
| Carbon monoxide | 2176.2 | $3.9 \times 10^{-3}$ | This work | 28 |
| Formaldehyde | 2778.4 | $3.6 \times 10^{-4}$ | Hanst (n.d.) | 0.8 |
| Hexane equivalent | 2900 | $6.0 \times 10^{-4}$ | Hanst (1971) | 13 |

### C. Oil Refinery

Large industrial complexes, such as oil refineries, are considered as "extended area" sources of pollutants. In other words, the pollutants from such complexes are generally a mixture of emissions from stacks, leaks, etc., and are classified as fugitive emissions. Determining the total emissions by conventional point monitors at such a complex source would be very difficult and time consuming, if possible at all. Thus, one of the primary goals of our laboratory remote sensing program is to develop the methodology for remote measurement of mass emission rates for extended area sources. This methodology would include remote measurement of wind velocity, as well as remote measurement of species concentrations (see Herget and Connor, 1977). In order to obtain some basic data on the ability of the ROSE system to measure emissions from such a source, a series of measurements were conducted at an oil refinery.

Fortunately, it was possible to make our initial measurements at a refinery situated in a valley. In this way a portion of the 1440 m optical path for absorption measurements was above the height of most of the refinery stacks. The site for the light source unit was reachable only by a four-wheel-drive vehicle, but power was available. The van was located on refinery property. The optical path was across the northern boundary of the refinery, so that winds with a southerly component would blow refinery emissions through the optical path.

Typical absorption spectra obtained at different times are shown in Figs. 10 and 11. The calculated concentrations are given in Table 2. The ambient temperature during these measurements was generally below freezing, so the water vapor concentration was much less than normally encountered. Because of the low temperatures and also the high winds, the quartz–iodine light source operated at cooler external temperature than in previous measurements, and the long wavelength spectra have a poorer signal-to-noise ratio than the short wavelength spectra. Although



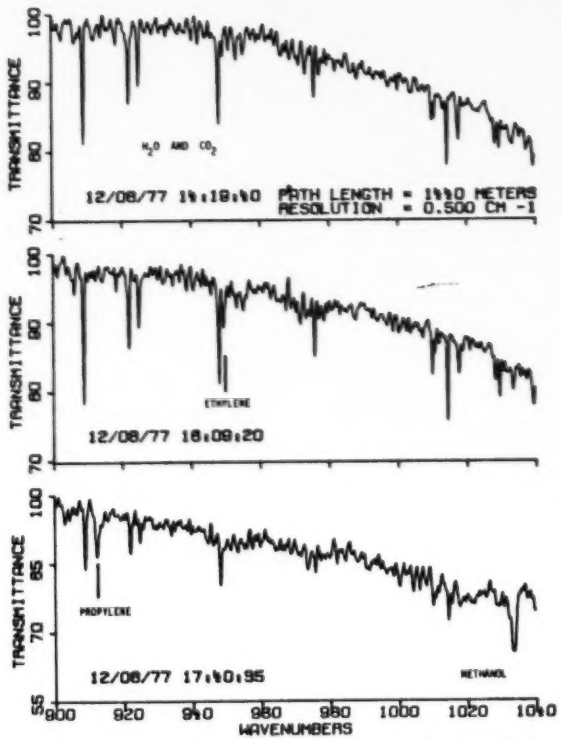


**Fig. 10.** Oil refinery absorption spectra (long wavelength region).

the $SO_2$ bands at 2500 $cm^{-1}$ is appreciably weaker than that at 1150 $cm^{-1}$, it is more suitable for concentration determination because there is essentially no interference from other species. It is of interest to note that the hydrocarbon continuum absorption in the 2800–3100 $cm^{-1}$ region has the same general shape, whether it be due to oil refinery emissions, jet engine or automobile engine emissions, or kerosene vapor. The strong continuum observed indicates that hydrocarbons other than ethylene, propylene, and methanol probably are present; time has not permitted analyses for other species. From similar spectra (not shown) the CO concentration was determined to be about 0.35 ppm, and methane concentrations about three times as great as ambient levels were observed.

The emission spectra obtained by sighting on two refinery stacks, a sulfur recovery unit and a CO boiler, are shown in Fig. 12. The sulfur recovery unit is characterized by strong CO and $SO_2$ emission, and weak

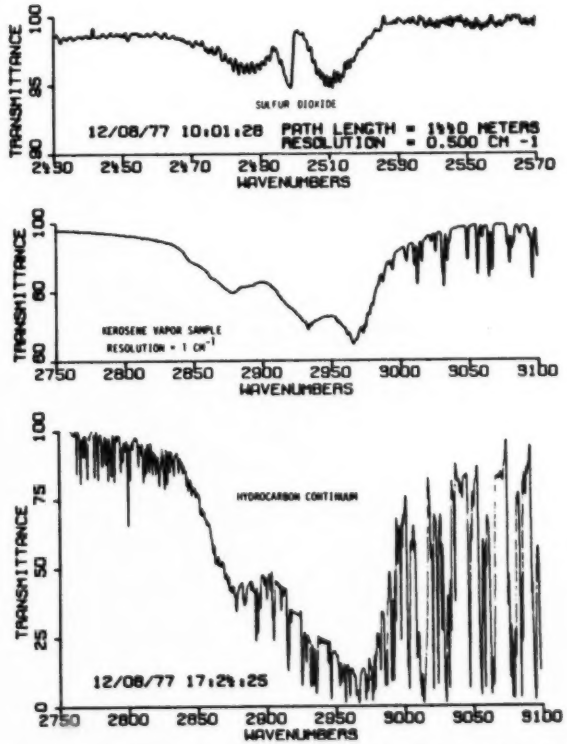


**Fig. 11.** Oil refinery absorption spectra (short wavelength region).

**TABLE 2**

Species Concentrations around an Oil Refinery (1440 m path)

| Species | $\nu$ ($cm^{-1}$) | $K(\nu)$ (ppm meter)$^{-1}$ | Reference for $K(\nu)$ | $C$ (ppm) |
|---|---|---|---|---|
| Propylene | 912.3 | $1.10 \times 10^{-3}$ | This work | 0.081 |
| Ethylene | 949.4 | $3.3 \times 10^{-3}$ | This work | 0.011 |
| Methanol | 1033.5 | $1.7 \times 10^{-3}$ | Hanst (1971) | 0.080 |
| Sulfur dioxide | 2500 | $3.5 \times 10^{-4}$ | This work | 1.0 |
| Hexane equivalent | 2900 | $6.0 \times 10^{-4}$ | Hanst (1971) | 0.86 |



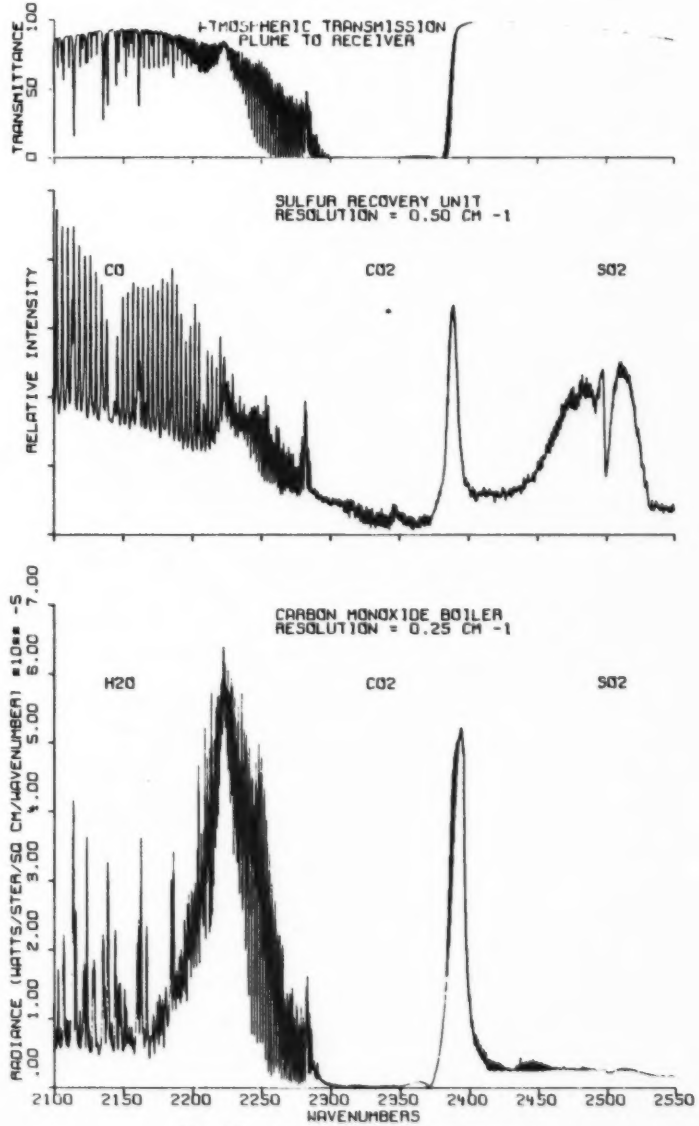


**Fig. 12.** Oil refinery emission spectra.

$CO_2$ emission; the intensity calibration on this spectrum is not meaningful, since the plume did not fill the instrument field of view. The CO boiler spectrum is characterized by strong $CO_2$ radiation and weak CO and $SO_2$ radiation. The $CO_2$ optical depth in this spectrum is sufficiently large that there is essentially blackbody radiation at 2225 $cm^{-1}$ where the cold atmospheric $CO_2$ does not absorb. This greatly simplifies a temperature calculation. The value obtained was 600°K, which is in good agreement with a nominal stack gas temperature of 650°K. Again, time has not permitted a complete analyses of these data.

## V. SUMMARY

A standard laboratory-model Fourier-transform spectrometer system has been installed in a van and used as a mobile instrument for obtaining the spectral signatures and concentrations of gaseous air pollutants by long-path absorption and remote emission spectroscopy. Of the various environments in which measurements have been made (rain storms, snow squalls, temperature extremes of −15° to 40°C, etc.) only the high vibration levels encountered in some of the jet engine measurements have adversely affected instrument operation. No particular problems have been encountered due to transportation of the interferometer system. After particularly cold nights, when the internal van temperature drops to near 0°C, it does require about an hour for the optics to stabilize when the van heating system is activated. Field data have been taken at 0.5, 0.25, and 0.125 $cm^{-1}$ spectral resolution. The lower resolution data were taken in some cases, where the particular species under study did not have resolvable fine structure, in order to save time and improve signal-to-noise ratios. However, it has been found advisable to collect data at 0.125 $cm^{-1}$ resolution in almost all cases since the effect of $H_2O$ interference is always minimized and the possibility of seeing unanticipated species is maximized by the higher resolution.

The accuracy of the long-path concentration measurements is considered to be better than ±10% (the principal source of error is determining the pressure in the calibration cell). There are a number of potential sources of error in determining a concentration solely from a remote emission measurement. We have not yet done sufficient work of this type, including comparison with in-stack data, to assess the measurement accuracy for this mode of operation.

The ROSE system has shown the capability of measuring concentrations as low as 0.01 ppm of various pollutants around several different types of sources. In the near future, it is planned to make additional measurements at an oil refinery, a pulp and paper mill, and on various types of



off-gas flares (where waste gases are burned at the top of a stack). Additional measurement configurations, in which the sun or the cold sky itself serves as the light source, will be evaluated. Although not discussed in this paper, a warm stack wall has already proven to be a valuable "light source" for absorption measurements; and more measurements of this type will be made. The ultimate purpose of these types of measurements is to be able to measure concentrations as a function of altitude. Thought has been given to locating light sources and/or a mirror on elevated platforms or towers, but we have not tried this as yet.

It is planned to make some measurements with the ROSE system in a cross-stack mode of operation. In this mode a collimated light beam will be transmitted through ports on opposite sides of a stack or duct, reflected off a flat mirror down to ground level, and into the van. This will allow absorption measurements to be made on stack gases for comparison with emission measurements made at the stack exit.

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